

# General form of magnetization damping: Magnetization dynamics of a spin system evolving nonadiabatically and out of equilibrium

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Using an effective Hamiltonian including the Zeeman and internal interactions, we describe the quantum theory of magnetization dynamics when the spin system evolves non-adiabatically and out of equilibrium. The Lewis-Riesenfeld dynamical invariant method is employed along with the Liouville-von Neumann equation for the density matrix. We derive a dynamical equation for magnetization defined with respect to the density operator with a general form of damping that involves the non-equilibrium contribution in addition to the Landau-Lifshitz-Gilbert equation. Two special cases of the radiation-spin interaction and the spin-spin exchange interaction are considered. For the radiation-spin interaction, the damping term is shown to be of the Gilbert type, while in the spin-spin exchange interaction case, the results depend on a coupled chain of correlation functions.

PACS numbers: 76.20.+q, 72.25.Ba

## I. INTRODUCTION

Magnetization dynamics in nanomagnets and thin films is rich in content, including such phenomena as giant magnetoresistance [1], spin-current-induced magnetization reversal [2],[3] and adiabatic spin pumping [4]. The study of the magnetization dynamics is motivated by theoretical interest in a deep and fundamental understanding of the physics of magnetic systems on a short time scale and out of equilibrium.

The results are important in gaining an understanding of the technological applications of magnetic systems to areas such as high-density memory and data storage devices. Recent results, in real time, provide the behavior of spins in particular set-ups of magnetic fields and measuring the spin flips as a function of time [5],[6].

The Landau-Lifshitz-Gilbert (LLG) equation [7],[8] provides a plausible phenomenological model for many experimental results. Recently, the LLG equation and the Gilbert damping term have been derived from an effective Hamiltonian including the radiation-spin interaction (RSI) [9]. It has been assumed there that the spin system maintains quasiadiabatic evolution.

However, a magnetic system whose Hamiltonian  $\hat{\mathcal{H}}(t)$  evolves nonadiabatically, i.e. in a non-equilibrium state, deviates far from quasi-equilibrium, and its density operator satisfies the quantum Liouville - von Neumann (LvN) equation

$$i\hbar \frac{\partial \hat{\rho}}{\partial t} + [\hat{\rho}, \hat{\mathcal{H}}]_- = 0. \quad (1)$$

In the present work, we aim to derive a magnetization equation including a damping term for such a nonequilibrium magnetic system. To find its nonadiabatic quantum states, we employ the Lewis-Riesenfeld (LR) dynamical invariant method [10]. This method originally designed for the nonequilibrium evolution of time-dependent quantum systems has been successfully applied to a variety of problems, including the nonadiabatic generalization of the Berry phase for the spin dynamics, nonequilibrium fermion systems etc [11],[12],[13]. In [14], the time-dependent invariants have been used for constructing the density operator for nonequilibrium systems. Following [14], we construct the density operator and then use it to determine the time evolution of magnetization.

The damping term represents magnetization relaxation processes due to the dissipation of magnetic energy. Various kinds of relaxation processes are usually melded together into a single damping term. Relativistic relaxation processes result in the Gilbert damping term with one damping parameter, while for the case of both exchange and relativistic relaxation the damping term is a tensor with several damping parameters [15], [16].

The relaxation processes are specified by interactions of spins with each other and with other constituents of the magnetic system. A derivation of the damping term from first principles should therefore start with a microscopic description of the interactions. Even though such microscopic derivation of damping has been performed for some relaxation processes (for instance, [17]), a full version of derivation for the Gilbert damping term has not yet been given, and in particular for a system in

non-equilibrium.

Herein, we consider a general spin system without specifying the interaction Hamiltonian and related relaxation processes. We start with a system of spins precessing in the effective magnetic field  $\vec{\mathbf{H}}_{eff}$  neglecting for a moment mutual interactions. Then at a fixed time later interactions in the system are switched on and influence the original precessional motion.

The interactions are assumed to be time-dependent, and the spin system evolves nonadiabatically out of equilibrium trying to relax to a new equilibrium magnetization. We perform a transformation, which is analogous to the one used in the transition to the interaction picture, to connect the density and magnetic moment operators before and after the time, when a new nonequilibrium dynamics starts, and to find an explicit expression for the interaction contribution to the magnetization equation.

Our paper is organized as follows. In Sec. II, the magnetization equation for the system of spins with a general form of interactions is derived. In Sec. III, the case of the radiation-spin interaction is considered, which is shown to produce the Gilbert damping term even for systems that are not in equilibrium. The contribution of the non-dissipative part of the radiation field to the magnetization equation and magnetization algebra is discussed. Sec. IV focuses on a special type of spin-spin interactions. We conclude with discussions in Sec. V.

## II. MAGNETIZATION EQUATION

Let us consider a quantum spin system defined by

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \lambda \hat{\mathcal{H}}_I, \quad (2)$$

where  $\hat{\mathcal{H}}_0$  is the Zeeman Hamiltonian describing the interaction of spins with an effective magnetic field

$$\hat{\mathcal{H}}_0 = -\gamma \sum_i \hat{S}_i \cdot \vec{\mathbf{H}}_{eff}(t), \quad (3)$$

$\gamma$  being the gyromagnetic ratio and  $\hat{S}_i$  being the spin operator of the  $i$ th atom, while the Hamiltonian  $\hat{\mathcal{H}}_I$  describes the internal interactions between the constituents of the spin system, including, for instance, the exchange and dipolar interactions between the atomic spins, as well as higher-order spin-spin interactions.

In Eq.(3), the effective magnetic field  $\vec{\mathbf{H}}_{eff}$  is given by the energy variational with magnetization,  $\vec{\mathbf{H}}_{eff} = -\delta E(M)/\delta \vec{\mathbf{M}}$ , where  $E(M)$  is the free energy of the magnetic system. This field includes the exchange field, the anisotropy field, and the demagnetizing field, as well as the external field,  $\vec{\mathbf{H}}_{ext}$ .

The interaction terms included in  $\hat{\mathcal{H}}_I$  are in general time-dependent, being switched on adiabatically or instantly at a fixed time  $t_0$ . The parameter  $\lambda$  in (2) can be chosen small in order to take into account the higher order effects perturbatively.

We introduce next the magnetic moment operator

$$\hat{\mathcal{M}} \equiv -\frac{\delta \hat{\mathcal{H}}}{\delta \vec{\mathbf{H}}_{ext}}, \quad (4)$$

which is the response of the spin system to the external field. The magnetization is defined as an ensemble average of the response

$$\vec{\mathbf{M}} = \langle \hat{\mathcal{M}} \rangle \equiv \frac{1}{V} \text{Tr}\{\hat{\rho} \hat{\mathcal{M}}\}, \quad (5)$$

where  $\hat{\rho}$  is the density operator satisfying the LvN equation (1) and  $V$  is the volume of the system. The explicit form of the density operator will be shown below.

For systems in equilibrium, the Hamiltonian itself satisfies the LvN equation and the density operator is expressed in terms of the Hamiltonian. For nonequilibrium systems, whose Hamiltonians are explicitly time dependent, the density operator is constructed by making use of the time-dependent adiabatic invariants.

### A. Zeeman precession

Let us first derive the magnetization equation for the Zeeman Hamiltonian. If the interactions,  $\hat{\mathcal{H}}_I$ , are switched off, the magnetic moment operator and the magnetization are

$$\hat{\mathcal{M}}_0 = -\frac{\delta \hat{\mathcal{H}}_0}{\delta \vec{\mathbf{H}}_{ext}} = \gamma \sum_i \hat{S}_i \quad (6)$$

and

$$\vec{\mathbf{M}}_0 = \langle \hat{\mathcal{M}}_0 \rangle_0 = \frac{\gamma}{V} \sum_i \text{Tr}\{\hat{\rho}_0 \hat{S}_i\}, \quad (7)$$

respectively, the operators  $\hat{\mathcal{M}}_0^a$ ,  $a = 1, 2, 3$ , fulfilling the  $SU(2)$  algebra

$$[\hat{\mathcal{M}}_0^a, \hat{\mathcal{M}}_0^b]_- = i\hbar\gamma\epsilon^{abc}\hat{\mathcal{M}}_0^c, \quad (8)$$

where the summation over repeated indices is assumed. In Eq.(7), the subscript "0" in the symbol  $\langle \dots \rangle_0$  indicates using of the density operator  $\hat{\rho}_0$ , which satisfies the equation

$$i\hbar \frac{\partial \hat{\rho}_0}{\partial t} + [\hat{\rho}_0, \hat{\mathcal{H}}_0]_- = 0. \quad (9)$$

Let  $\hat{\mathcal{I}}_0(t)$  be a non-trivial Hermitian operator, which is a dynamical invariant. That is,  $\hat{\mathcal{I}}_0(t)$  satisfies the LvN equation

$$\frac{d\hat{\mathcal{I}}_0}{dt} \equiv \frac{\partial \hat{\mathcal{I}}_0}{\partial t} + \frac{1}{i\hbar} [\hat{\mathcal{I}}_0, \hat{\mathcal{H}}_0]_- = 0. \quad (10)$$

As shown in [10], the eigenstates of  $\hat{\mathcal{I}}_0(t)$  can be used for evaluating the exact quantum states that are solutions

of the Schrödinger equation. The linearity of the LvN equation allows us to state that any analytic functional of  $\hat{\mathcal{I}}_0(t)$  satisfies the LvN equation provided that  $\hat{\mathcal{I}}_0(t)$  satisfies the same equation. In particular, we can use  $\hat{\mathcal{I}}_0(t)$  to define the density operator  $\hat{\rho}_0$  as [14]

$$\hat{\rho}_0(t) = \frac{1}{\mathcal{Z}_0} e^{-\beta \hat{\mathcal{I}}_0(t)}, \quad \mathcal{Z}_0 = \text{Tr}\{e^{-\beta \hat{\mathcal{I}}_0(t)}\}. \quad (11)$$

Here  $\beta$  is a free parameter and will be identified with the inverse temperature for the equilibrium system.

The LvN equation for  $\hat{\mathcal{I}}_0$  is formally solved by

$$\hat{\mathcal{I}}_0(t) = \hat{U}(t, t_0) \hat{\mathcal{I}}_0(t_0) \hat{U}(t_0, t), \quad (12)$$

where

$$\hat{U}(t_0, t) \equiv T \exp \left\{ \frac{i}{\hbar} \int_{t_0}^t d\tau \hat{\mathcal{H}}_0(\tau) \right\}, \quad (13)$$

and  $T$  denotes the time-ordering operator.

As the Zeeman Hamiltonian is linear in spin operators, we can take  $\hat{\mathcal{I}}_0(t)$  of the same form [18]

$$\hat{\mathcal{I}}_0(t) = \sum_i \hat{S}_i \cdot \vec{\mathbf{R}}_0(t), \quad (14)$$

where  $\vec{\mathbf{R}}_0$  is a vector parameter to be determined by a dynamical equation. Then, Eq.(9) becomes

$$\sum_i \hat{S}_i \cdot \left( \frac{d\vec{\mathbf{R}}_0}{dt} - \gamma \vec{\mathbf{R}}_0 \times \vec{\mathbf{H}}_{eff} \right) = 0 \quad (15)$$

and we obtain an equation for the vector parameter given by

$$\frac{d\vec{\mathbf{R}}_0}{dt} = -|\gamma| \vec{\mathbf{R}}_0 \times \vec{\mathbf{H}}_{eff}. \quad (16)$$

This equation explicitly describes the vector precessing with respect to the field  $\vec{\mathbf{H}}_{eff}$ . Thus, without loss of generality, we can identify  $\vec{\mathbf{R}}_0$  with the magnetization vector  $\vec{\mathbf{M}}_0$  (up to a dimensional constant factor) and Eq.(16) with the equation of motion of magnetization for the Zeeman Hamiltonian  $\hat{\mathcal{H}}_0$ .

An alternative method to obtain the magnetization equation is just to differentiate both sides of Eq.(7) with respect to time and use Eq.(9). In this way, we arrive at Eq.(16) with  $\vec{\mathbf{R}}_0 = \vec{\mathbf{M}}_0$ .

## B. Interactions and damping

In the case, when the interactions are present, the density operator for the full Hamiltonian may be written as

$$\hat{\rho}(t) = \frac{1}{\mathcal{Z}} e^{-\beta \hat{\mathcal{I}}(t)}, \quad \mathcal{Z} = \text{Tr}\{e^{-\beta \hat{\mathcal{I}}(t)}\}, \quad (17)$$

where  $\hat{\mathcal{I}}$  is the invariant operator for the system. As the interaction Hamiltonian is in general non-linear in

spin operators,  $\hat{\mathcal{I}}(t)$  cannot be taken in the form given by Eq.(14). Moreover, the form of  $\hat{\mathcal{I}}(t)$  can not be determined without specifying the interactions.

To derive the magnetization equation in this case, we proceed as follows. We perform, on  $\hat{\rho}(t)$ , the transformation defined by the operator (13),

$$\hat{\rho} \rightarrow \hat{\rho}_{int} \equiv \hat{U}(t_0, t) \hat{\rho}(t) \hat{U}(t, t_0), \quad (18)$$

removing the Zeeman interaction. For systems with  $\hat{\mathcal{H}}_0$  constant in time, the operator  $\hat{U}(t_0, t) = \exp\{(i/\hbar)\hat{\mathcal{H}}_0(t-t_0)\}$  leads to the interaction picture, which proves to be very useful for all forms of interactions since it distinguishes among the interaction times. For our system with both  $\hat{\mathcal{H}}_0$  and  $\hat{\mathcal{H}}_I$  dependent on time, the operator (13) plays the same role, removing the unperturbed part of the Hamiltonian from the LvN equation.

Substituting Eq.(18) into (1), yields

$$i\hbar \frac{\partial \hat{\rho}_{int}}{\partial t} = \lambda [\hat{\mathcal{H}}_{int}, \hat{\rho}_{int}]_-, \quad (19)$$

where

$$\hat{\mathcal{H}}_{int}(t) \equiv \hat{U}(t_0, t) \hat{\mathcal{H}}_I(t) \hat{U}(t, t_0). \quad (20)$$

The magnetic moment operator and the magnetization become

$$\hat{\mathcal{M}} = \hat{\mathcal{M}}_0 + \hat{\mathcal{M}}_I, \quad (21)$$

where

$$\hat{\mathcal{M}}_I \equiv -\lambda \frac{\delta \hat{\mathcal{H}}_I}{\delta \vec{\mathbf{H}}_{ext}}, \quad (22)$$

and

$$\vec{\mathbf{M}} = \frac{1}{V} \text{Tr}\{\hat{\rho}_{int}(\hat{\mathcal{M}}_{0,int} + \hat{\mathcal{M}}_{I,int})\}, \quad (23)$$

$\hat{\mathcal{M}}_{0,int}$  and  $\hat{\mathcal{M}}_{I,int}$  being related with  $\hat{\mathcal{M}}_0$  and  $\hat{\mathcal{M}}_I$ , respectively, in the same way as  $\hat{\mathcal{H}}_{int}$  is related with  $\hat{\mathcal{H}}_I$ .

The operators  $\hat{\mathcal{M}}_{0,int}$ ,  $\hat{\mathcal{M}}_{I,int}$  are generally used to calculate the magnetic susceptibility [19]. Let us show now how these operators determine the time evolution of magnetization. The evolution in time of  $\hat{\mathcal{M}}_{0,int}$  is given by the equation

$$\begin{aligned} \frac{\partial \hat{\mathcal{M}}_{0,int}}{\partial t} &= \frac{i}{\hbar} \hat{U}(t_0, t) [\hat{\mathcal{H}}_0, \hat{\mathcal{M}}_0]_- \hat{U}(t, t_0) \\ &= \gamma \hat{\mathcal{M}}_{0,int} \times \vec{\mathbf{H}}_{eff}, \end{aligned} \quad (24)$$

which is analogous to Eq.(16). It describes the magnetization precessional motion with respect to  $\vec{\mathbf{H}}_{eff}$ .

The equation for  $\hat{\mathcal{M}}_{I,int}$ ,

$$\frac{\partial \hat{\mathcal{M}}_{I,int}}{\partial t} = \frac{i}{\hbar} \hat{U}(t_0, t) [\hat{\mathcal{H}}_0, \hat{\mathcal{M}}_I]_- \hat{U}(t, t_0)$$

$$+ \hat{U}(t_0, t) \frac{\partial \hat{\mathcal{M}}_I}{\partial t} \hat{U}(t, t_0) \quad (25)$$

describes more complex magnetization dynamics governed by the interaction Hamiltonian  $\hat{\mathcal{H}}_I$ . In addition,  $\hat{\mathcal{M}}_I$  depends on time explicitly. However, this dynamics includes the precessional motion as well, since the interactions induced magnetization is a part of the precessing total magnetization  $\vec{\mathbf{M}}$ . Introducing

$$\vec{\mathbf{D}}_I \equiv \frac{i}{\hbar} [\hat{\mathcal{H}}_0, \hat{\mathcal{M}}_I]_- - \gamma \hat{\mathcal{M}}_I \times \vec{\mathbf{H}}_{eff} \quad (26)$$

to represent deviations from the purely precessional motion, we bring the equation for  $\hat{\mathcal{M}}_{I,int}$  into the following form

$$\begin{aligned} \frac{\partial \hat{\mathcal{M}}_{I,int}}{\partial t} &= \gamma \hat{\mathcal{M}}_{I,int} \times \vec{\mathbf{H}}_{eff} \\ &+ \hat{U}(t_0, t) \left( \frac{\partial \hat{\mathcal{M}}_I}{\partial t} + \vec{\mathbf{D}}_I \right) \hat{U}(t, t_0). \end{aligned} \quad (27)$$

Taking the time-derivative of  $\vec{\mathbf{M}}$  given by Eq.(23) and using Eq.(19), we obtain

$$\begin{aligned} \frac{d\vec{\mathbf{M}}}{dt} &= \frac{1}{V} \text{Tr} \left\{ \hat{\rho}_{int} \left( \frac{\partial \hat{\mathcal{M}}_{0,int}}{\partial t} + \frac{\partial \hat{\mathcal{M}}_{I,int}}{\partial t} \right) \right. \\ &\quad \left. + \frac{\lambda}{i\hbar} [\hat{\mathcal{M}}_{0,int} + \hat{\mathcal{M}}_{I,int}, \hat{\mathcal{H}}_{int}]_- \right\}. \end{aligned} \quad (28)$$

Substituting next Eqs.(24) and (27) into Eq.(28), finally yields

$$\frac{d\vec{\mathbf{M}}}{dt} = -|\gamma| \vec{\mathbf{M}} \times \vec{\mathbf{H}}_{eff} + \vec{\mathbf{D}}, \quad (29)$$

where

$$\vec{\mathbf{D}} \equiv \lambda \left\langle \frac{1}{i\hbar} [\hat{\mathcal{M}}, \hat{\mathcal{H}}_I]_- \right\rangle + \left\langle \frac{\partial \hat{\mathcal{M}}_I}{\partial t} + \vec{\mathbf{D}}_I \right\rangle. \quad (30)$$

Therefore, Eq.(29) is the magnetization equation for the system specified by (2). This equation is general since it is derived without specifying  $\hat{\mathcal{H}}_I$ . The  $\vec{\mathbf{D}}$ -term contains all effects that the interactions,  $\hat{\mathcal{H}}_I$ , can have on the magnetization precession, so that Eq.(29) is complete.

The contribution of  $\hat{\mathcal{H}}_I$  to the  $\vec{\mathbf{D}}$ -term in the magnetization equation can be divided into two parts. One is proportional to  $\langle [\hat{\mathcal{M}}, \hat{\mathcal{H}}_I]_- \rangle$  and is related to the change in the density matrix when the interactions of  $\hat{\mathcal{H}}_I$  are switched on. The second part  $\langle \frac{\partial \hat{\mathcal{M}}_I}{\partial t} + \vec{\mathbf{D}}_I \rangle$  originates from the change in the magnetization itself. Which part of  $\vec{\mathbf{D}}$  is dominating depends on the nature of the interactions.

### III. RADIATION-SPIN INTERACTION

One of the important issues in the study of the magnetization dynamics is the relaxation phenomena. The magnetization relaxation mechanism can be introduced by various interactions such as spin-orbit coupling and two-magnon scattering. In this section, we calculate the  $\vec{\mathbf{D}}$ -term for the magnetization relaxation process, which is induced by the RSI.

#### A. Dissipative radiation field

The RSI approach is an effective field method, in which contributions to the magnetization relaxation are effectively represented by the radiation-spin interaction [9]. In this method, the damping imposed on the precessing magnetization originates from the magnetization precessional motion itself. It is assumed that a radiation field is induced by the precession and that this field acts back on the magnetization producing a dissipative torque.

The Hamiltonian for the RSI is

$$\lambda \hat{\mathcal{H}}_I = -\gamma \sum_i \hat{S}_i \cdot \vec{\mathbf{H}}_r^d, \quad (31)$$

where

$$\vec{\mathbf{H}}_r^d \equiv \lambda (\vec{\mathbf{M}} \times \vec{\mathbf{H}}_{eff} - \alpha M^2 \vec{\mathbf{H}}_{eff}) \quad (32)$$

is the dissipative part of the effective radiation field, i.e. the part responsible for the dissipative torque, and  $M$  is the magnitude of magnetization. The parameter  $\alpha$  will be specified below, while  $\lambda$  in Eq.(2) is now the radiation parameter.

The RSI contribution to the magnetic moment is

$$\hat{\mathcal{M}}_{I,d} = -\lambda (\alpha M^2 \hat{\mathcal{M}}_0 - \hat{\mathcal{M}}_0 \times \vec{\mathbf{M}}), \quad (33)$$

and the total magnetization becomes

$$\vec{\mathbf{M}} = (1 - \lambda \alpha M^2) \langle \hat{\mathcal{M}}_0 \rangle + \lambda \langle \hat{\mathcal{M}}_0 \rangle \times \vec{\mathbf{M}}. \quad (34)$$

It turns out that the vectors  $\vec{\mathbf{M}}$  and  $\langle \hat{\mathcal{M}}_0 \rangle$  are parallel. Indeed, taking the vector product of both sides of Eq.(34) with  $\langle \hat{\mathcal{M}}_0 \rangle$ , yields the equation

$$\begin{aligned} \langle \hat{\mathcal{M}}_0 \rangle \times \vec{\mathbf{M}} &= \lambda \langle \hat{\mathcal{M}}_0 \rangle \times \langle \hat{\mathcal{M}}_0 \rangle \times \vec{\mathbf{M}} \\ &= \lambda \langle \hat{\mathcal{M}}_0 \rangle^2 \left[ (1 - \lambda \alpha M^2) \langle \hat{\mathcal{M}}_0 \rangle - \vec{\mathbf{M}} \right], \end{aligned} \quad (35)$$

which is valid only if  $\vec{\mathbf{M}}$  is parallel to  $\langle \hat{\mathcal{M}}_0 \rangle$ , so that

$$\langle \hat{\mathcal{M}}_0 \rangle = \frac{\vec{\mathbf{M}}}{1 - \lambda \alpha M^2}. \quad (36)$$

A straightforward calculation shows that

$$\langle \vec{\mathbf{D}}_I \rangle = -\lambda \left\langle \frac{1}{i\hbar} [\hat{\mathcal{M}}, \hat{\mathcal{H}}_I]_- \right\rangle$$

$$= -\frac{\gamma\lambda}{1-\lambda\alpha M^2}\vec{M} \times \vec{M} \times \vec{H}_{eff}, \quad (37)$$

so that the contributions of  $\vec{D}_I$  and  $[\hat{\mathcal{M}}, \hat{H}_I]_-$  to Eq.(30) cancel each other. Calculating next the time derivative of  $\hat{\mathcal{M}}_{I,d}$  and using Eq.(36), we obtain

$$\vec{D} = \alpha\vec{M} \times \frac{d\vec{M}}{dt}, \quad (38)$$

provided the following relation between the parameters  $\alpha$  and  $\lambda$  holds

$$\alpha = \frac{\lambda}{1-\lambda\alpha M^2}. \quad (39)$$

Therefore, Eq.(29) takes the form of the LLG equation,  $\alpha$  becoming a damping parameter.

The relation given by Eq.(39) reflects the origin of the radiation field. Both the radiation and damping parameters depend on the magnetization. The dimensionless parameters independent of  $M$  are  $\lambda_0 \equiv \lambda M$  and  $\alpha_0 \equiv \alpha M$  with the relation

$$\lambda_0 = \frac{\alpha_0}{1+\alpha_0^2}. \quad (40)$$

The equivalent form of the LLG equation, which is more suitable for calculations, is

$$\frac{d\vec{M}}{dt} = -|\gamma|\vec{M} \times \left[ (1-\lambda\alpha M^2)\vec{H}_{eff} + \lambda\vec{M} \times \vec{H}_{eff} \right]. \quad (41)$$

Let us assume that  $\vec{H}_{eff}$  is a uniform static field in the  $z$ -direction, i.e.  $\vec{H}_{eff} = (0, 0, H_z)$ . Then the magnetization equation becomes, in component form,

$$\frac{d}{dt}M_p^2 = -2\lambda\omega_0 M_z M_p^2, \quad (42)$$

$$\frac{d}{dt}M_z = \lambda\omega_0 M_p, \quad (43)$$

where  $\omega_0 \equiv |\gamma|H_z$  is the frequency of magnetization precession and  $M_p^2 \equiv M_x^2 + M_y^2 = M^2 - M_z^2$ . These equations are solved exactly by

$$M_z = M \cdot \frac{\tanh[\lambda_0\omega_0(t-t_0)] + d}{1 + d \cdot \tanh[\lambda_0\omega_0(t-t_0)]} \quad (44)$$

and

$$M_p = \frac{M\sqrt{1-d^2}}{\cosh[\lambda_0\omega_0(t-t_0)]} \cdot \frac{1}{1 + d \cdot \tanh[\lambda_0\omega_0(t-t_0)]}, \quad (45)$$

where  $d$  stands for the initial condition

$$d \equiv \frac{M_z(t=t_0)}{M}.$$

In the limit  $t \rightarrow \infty$ ,  $M_z$  tends to  $M$  and  $M_p$  tends to zero, so that during the relaxation process the magnetization vector tends to be parallel to the effective magnetic field, and the relaxation characteristic time  $\tau$  is  $1/(\lambda_0\omega_0)$ .

## B. General radiation field

The RSI approach can be directly used for studying the effect of the real radiation-spin interaction in the magnetization relaxation process. In that case, one has to distinguish the real radiation field contribution to the damping parameter from the effective one. It is therefore important to consider possible generalizations of the ansatz given by Eq.(32) to get a more detailed picture of the RSI.

The dissipative radiation field in Eq.(32) is a composite of two fields, which are parallel and perpendicular to  $\vec{H}_{eff}$ , respectively. The field parallel to  $\vec{H}_{eff}$  changes only the frequency of the magnetization precessional motion, while the torque  $\vec{H}_{eff} \times \vec{M}$  introduces a damping effect as well.

The choice of the field  $\vec{H}_r^d$  made in Eq.(32) is not a general one. Let us modify it by applying additional fields and see how the parameters in the magnetization equation are changed. If we apply an additional field in the direction of  $\vec{H}_{eff}$  and modify  $\vec{H}_r^d$  as

$$\vec{H}_r^d \rightarrow \vec{H}_r^d - \bar{\lambda}_0 \vec{H}_{eff}, \quad (46)$$

where  $\bar{\lambda}_0$  is an arbitrary dimensionless parameter, then this results in a change of the magnitude of the effective magnetic field and therefore in the change of the frequency of the magnetization precession as

$$\omega_0 \rightarrow \omega \equiv \omega_0(1 - \bar{\lambda}_0). \quad (47)$$

For  $\bar{\lambda}_0 > 1$ , the magnetization vector turns upside down and the precession continues in an opposite direction.

The magnetization damping is not affected by the additional field. Rescaling of  $\vec{H}_{eff}$  in  $\vec{H}_r^d$  leads to a change of the radiation parameter as well,

$$\lambda_0 \rightarrow \frac{1}{1 - \bar{\lambda}_0} \lambda_0, \quad (48)$$

so that the relaxation characteristic time remains the same.

If we apply an additional torque,

$$\vec{H}_r^d \rightarrow \vec{H}_r^d + \bar{\lambda}_0 \vec{M} \times \vec{H}_{eff}, \quad (49)$$

then the parameters in the ansatz (32) change as follows

$$\lambda_0 \rightarrow \lambda_0 + \bar{\lambda}_0, \quad (50)$$

$$\alpha_0 \rightarrow \frac{\lambda_0}{\lambda_0 + \bar{\lambda}_0} \alpha_0, \quad (51)$$

and the relaxation characteristic time becomes

$$\tau \rightarrow \bar{\tau} \equiv \tau \left( 1 + \frac{\bar{\lambda}_0}{\lambda_0} \right)^{-1}, \quad (52)$$

decreasing for  $\bar{\lambda}_0 > 0$  and increasing for  $-\lambda_0 < \bar{\lambda}_0 < 0$ . If the additional torque is stronger than the original one

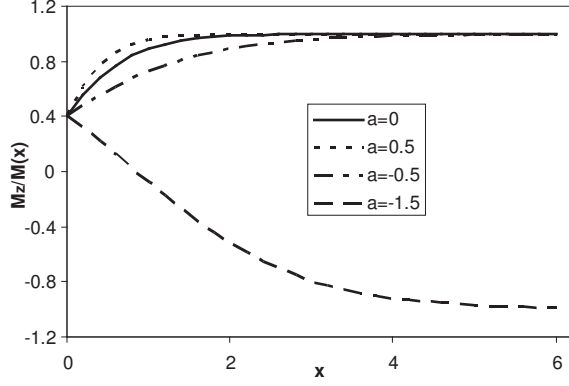


FIG. 1: The dependence of the  $z$ -component of the magnetization vector,  $M_z/M$ , on time  $x = (t - t_0)/\tau$  is shown for different values of  $a$  and for  $d = 0.4$ .

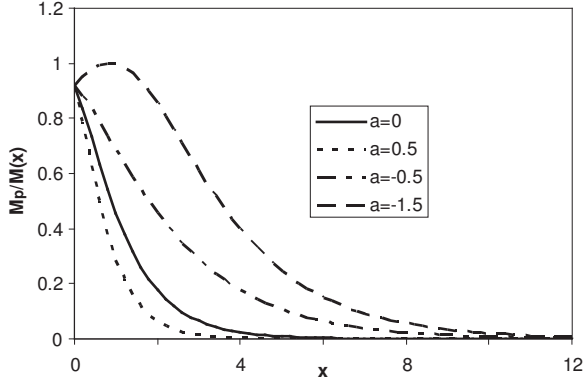


FIG. 2: The time dependence of the perpendicular component of the magnetization vector,  $M_p/M$ , is plotted for different values of  $a$  and for  $d = 0.4$ .

and in the opposite direction, i.e.  $\bar{\lambda}_0 < -\lambda_0$ , then the magnetization vector turns over again. For  $\bar{\lambda}_0 = -\lambda_0$ , the additional and original torques cancel each other, and there is no damping.

In Fig.1 and Fig.2 we plot the solutions for  $M_z$  and  $M_p$  given by Eq.(44) and Eq.(45), respectively, in the presence of an additional torque (nonzero  $a$ 's) and without it ( $a = 0$ ), where  $a \equiv \bar{\lambda}_0/\lambda_0$ . We observe a change in the relaxation time depending on the sign of  $a$ . For  $a > 0$ , the additional torque is in the same direction as the original one, while for  $a < 0$  their directions are opposite. A drastic change occurs for  $a < -1$ , when  $M_z$  tends to  $(-M)$  indicating the overturning of  $\vec{M}$ . The magnetization vector first becomes perpendicular to  $\vec{H}_{eff}$ , when  $M_p/M$  reaches its maximum value, and then it turns up-side down.

The radiation field  $\vec{H}_r$  can contain a non-dissipative part as well, i.e.  $\vec{H}_r = \vec{H}_r^d + \vec{H}_r^n$ . Let us assume that  $\vec{H}_r^n$

is parallel to  $\vec{M}$  and we take it of the form

$$\vec{H}_r^n = \kappa \lambda \frac{\vec{M}}{M} (\vec{M} \cdot \vec{H}_{eff}), \quad (53)$$

where  $\kappa$  is an arbitrary parameter.

The field given by Eq.(53) does not change the magnetization precessional motion. However, the magnetic moment operator is given as

$$\hat{\mathcal{M}} \rightarrow \hat{\mathcal{M}} + \hat{\mathcal{M}}_{I,n} \equiv \hat{\mathcal{M}} + \kappa \lambda \frac{\vec{M}}{M} (\hat{\mathcal{M}}_0 \cdot \vec{M}). \quad (54)$$

The relation between the vectors  $\langle \hat{\mathcal{M}}_0 \rangle$  and  $\vec{M}$  becomes

$$\langle \hat{\mathcal{M}}_0 \rangle = \frac{\vec{M}}{1 - \lambda(\alpha M + \kappa)M}. \quad (55)$$

Proceeding in the same way as before, when the non-dissipative part of the radiation field was omitted, to calculate the total radiation field contribution to the  $\vec{D}$ -term, we obtain the same Gilbert-type structure of the damping term with the same damping parameter, i.e. the part of the radiation field that is parallel to  $\vec{M}$  does not contribute to the magnetization equation.

### C. Magnetization algebra

The magnetization algebra, i.e. the algebra of magnetic moment operators, gives a further insight into the RSI. Let us study how the non-dissipative part of the radiation field contributes to this algebra.

Although we can omit the non-dissipative radiation field in the procedure obtaining the equation of motion for magnetization, it is important to take this field into account when we construct the magnetization algebra. Indeed, the operators  $\hat{\mathcal{M}}^a = \hat{\mathcal{M}}_0^a + \hat{\mathcal{M}}_{I,d}^a$  without the non-dissipative radiation field contribution fulfil the algebra

$$\begin{aligned} [\hat{\mathcal{M}}^a, \hat{\mathcal{M}}^b]_- &= i\hbar\gamma\epsilon^{abc}\{(1 - \lambda\alpha M^2) \cdot \hat{\mathcal{M}}^c \\ &+ \frac{1}{\kappa}\lambda M \cdot \hat{\mathcal{M}}_{I,n}^c\}, \end{aligned} \quad (56)$$

which, however, contains  $\hat{\mathcal{M}}_{I,n}$  on its right-hand side and therefore the algebra is not closed.

Let us define, as in Eq.(54), the total magnetic moment operator  $\hat{\mathcal{M}}_{tot} \equiv \hat{\mathcal{M}} + \hat{\mathcal{M}}_{I,n}$ . Then the magnetization algebra becomes

$$\begin{aligned} [\hat{\mathcal{M}}_{tot}^a, \hat{\mathcal{M}}_{tot}^b]_- &= i\hbar\gamma\epsilon^{abc}\{(1 - \lambda\alpha M^2 + \kappa\lambda M) \cdot \hat{\mathcal{M}}^c \\ &+ (\frac{1}{\kappa}\lambda M - 1 + \lambda\alpha M^2) \cdot \hat{\mathcal{M}}_{I,n}^c\}. \end{aligned} \quad (57)$$

Fixing next the parameter  $\kappa$  by taking it as a solution of the equation

$$\frac{1}{\kappa} - \kappa = \frac{2}{\alpha_0}, \quad (58)$$

that is

$$\kappa_{\pm} = -\frac{1}{\alpha_0}(1 \mp \sqrt{1 + \alpha_0^2}), \quad (59)$$

brings the algebra into the closed, standard form for  $SU(2)$  algebra

$$[\hat{\mathcal{M}}_{tot}^a, \hat{\mathcal{M}}_{tot}^b]_{-} = i\hbar\gamma_{tot}\varepsilon^{abc}\hat{\mathcal{M}}_{tot}^c, \quad (60)$$

where

$$\gamma_{tot} \equiv \pm\gamma \frac{1}{\sqrt{1 + \alpha_0^2}}. \quad (61)$$

For  $0 < \alpha_0 < 1$ ,  $\kappa_{+} > 0$  and  $\kappa_{-} < 0$ , the sign (+) in Eq.(61) corresponding to the case  $\kappa = \kappa_{+}$  and sign (-) to  $\kappa = \kappa_{-}$ .

Therefore, the RSI preserves the form of the magnetization algebra (c.f. Eq.(8)), by renormalizing the gyromagnetic ratio, only if the interaction of spins with the non-dissipative part of the radiation field is included in a proper way. If the direction of  $\vec{\mathbf{H}}_r^n$  is the same as that of  $\vec{\mathbf{M}}$  and  $\kappa = \kappa_{+}$ , the gyromagnetic ratio remains negative, while its absolute value decreases. If the direction of  $\vec{\mathbf{H}}_r^n$  is opposite to that of  $\vec{\mathbf{M}}$  and  $\kappa = \kappa_{-}$ , then the gyromagnetic ratio changes its sign and becomes positive.

#### IV. SPIN-SPIN INTERACTIONS

The spin-spin interactions among the spins in the system introduce many body effects, which can be treated perturbatively in the weak coupling regime. In this case the  $\vec{\mathbf{D}}$ -term can be expanded in powers of  $\lambda$ . To demonstrate this, we consider the spin-spin interactions of a specific type. The interaction between spins is usually an exchange interaction of the form

$$-2J \sum_{i,j} \hat{S}_i \hat{S}_j = -\frac{2J}{\gamma^2} \hat{\mathcal{M}}_0^2, \quad (62)$$

the coupling constant  $J$  being called the exchange integral. We generalize the ansatz given by Eq.(62) by assuming that the exchange integral depends on the magnetization and introduce the spin-spin interactions as follows

$$\lambda \hat{\mathcal{H}}_I = \sum_{i,j} J^{ab}(M) \hat{S}_i^a \hat{S}_j^b, \quad (63)$$

where  $J^{ab} = \lambda M^a M^b$ . Since  $\hat{\mathcal{H}}_I$  does not depend explicitly on the external field, its contribution to the magnetic moment operator vanishes,  $\hat{\mathcal{M}}_I = 0$ .

The non-vanishing commutator  $[\hat{\mathcal{M}}_0, \hat{\mathcal{H}}_I]_{-}$  in Eq.(30) is the only contribution of the spin-spin interaction to the magnetization equation, resulting in

$$\vec{\mathbf{D}} = \frac{\lambda}{\gamma} \vec{\mathbf{M}} \times \vec{\mathbf{\Omega}}, \quad (64)$$

where

$$\Omega^a \equiv \langle [\hat{\mathcal{M}}_0^a, \hat{\mathcal{M}}_0^b]_{+} \rangle M^b, \quad (65)$$

and

$$[\hat{\mathcal{M}}_0^a, \hat{\mathcal{M}}_0^b]_{+} \equiv \hat{\mathcal{M}}_0^a \hat{\mathcal{M}}_0^b + \hat{\mathcal{M}}_0^b \hat{\mathcal{M}}_0^a. \quad (66)$$

The correlation function  $G^{ab} \equiv \langle [\hat{\mathcal{M}}_0^a, \hat{\mathcal{M}}_0^b]_{+} \rangle$  is the sum of spin correlation functions,

$$G^{ab} = 2\gamma^2 \sum_i \sum_{j \neq i} \langle \hat{S}_i^a \hat{S}_j^b \rangle, \quad (67)$$

excluding the self-interaction of spins. For the standard ansatz given in Eq.(62),  $\vec{\mathbf{D}} = 0$  and the magnetization equation does not change.

If the spin-spin interactions are turned on at  $t = t_0$ , so that  $\hat{\rho}(t_0) = \hat{\rho}_0(t_0)$ , then, integrating both sides of Eq.(19), we find

$$\hat{\rho}_{\lambda}(t) = \hat{\rho}_0(t_0) + \frac{\lambda}{i\hbar} \int_{t_0}^t d\tau [\hat{\mathcal{H}}_{\lambda}(\tau), \hat{\rho}_{\lambda}(\tau)]_{-}. \quad (68)$$

Substituting Eq.(68) into the definition of  $G^{ab}$ , yields the equation

$$G^{ab}(t) = G_0^{ab}(t_0) + \frac{1}{\gamma} \int_{t_0}^t d\tau J^{cd}(\tau) \left( \varepsilon^{ace} G^{ebd}(\tau) + \varepsilon^{bce} G^{aed}(\tau) \right), \quad (69)$$

where

$$G_0^{ab} \equiv \langle [\hat{\mathcal{M}}_0^a, \hat{\mathcal{M}}_0^b]_{+} \rangle_0, \quad (70)$$

which relates  $G^{ab}$  to the third order correlation function, i.e. the correlation function of the product of three magnetic moment operators,

$$G^{abc} \equiv \langle [\hat{\mathcal{M}}_0^a, \hat{\mathcal{M}}_0^b]_{+}, \hat{\mathcal{M}}_0^c \rangle_{+}. \quad (71)$$

The correlation function  $G^{abc}$ , in turn, is related to the fourth order correlation function and etc., and we have therefore an infinite number of coupled equations for spin correlation functions. For any practical calculation this infinite hierarchy has to be truncated. That then defines the approximation scheme which may be considered on the basis of the physical requirements for the system. The approximation scheme will depend on the physical

properties such as density and on the strength of the interactions.

If the Hamiltonian  $\hat{\mathcal{H}}_I$  is a small perturbation to the original  $\hat{\mathcal{H}}_0$ , we can solve Eq.(68) perturbatively. In the lowest, zeroth order in  $\lambda$ , we replace  $\hat{\rho}_\lambda(t)$  by  $\hat{\rho}_0(t_0)$ , so that  $G^{ab} \approx G_0^{ab}(t_0)$ . We choose the initial value for  $G^{ab}$  as

$$\sum_i \sum_{j \neq i} \langle \hat{S}_i^a \hat{S}_j^b \rangle_0 = I^{ab} \quad (72)$$

with  $I^{xx} = I^{yy} = 0$ ,  $I^{zz} = I$  and  $I^{ab} = 0$  for  $a \neq b$ . We also define again the  $z$ -direction as the direction of the effective magnetic field that is chosen uniform and static. Then the  $\vec{\mathbf{D}}$ -term becomes, in component form,

$$D_x = 2\lambda\gamma IM_y M_z, \quad (73)$$

$$D_y = -2\lambda\gamma IM_x M_z, \quad (74)$$

$$D_z = 0. \quad (75)$$

producing two effects in the magnetization equation: the magnetization is now precessing with respect to  $(H_z - 2\lambda IM_z)$ , its  $z$ -component remaining constant in time,  $(d/dt)M_z = 0$ , and the frequency of the precession is

$$\bar{\omega}_0 \equiv \omega_0 \left( 1 - \lambda \frac{2IM_z}{H_z} \right). \quad (76)$$

Therefore, in the lowest order of perturbations, when the  $\vec{\mathbf{D}}$  is linear in  $\lambda$ , it shifts the direction and the frequency of the precessional motion without introducing damping effects. To find a role of the higher powers of  $\lambda$  in  $\vec{\mathbf{D}}$  and to determine how they affect the magnetization equation, a truncation of the chain of spin correlation equations is needed. This would require a consistent perturbation approach to the hierarchy of the coupled equations for the correlation functions.

## V. DISCUSSIONS

We have derived a general form of magnetization equation for a system of spins precessing in an effective magnetic field without specifying the internal interactions. It can be applied in the study of magnetization dynamics of any type, including nonequilibrium and nonlinear effects, provided the interaction of individual spins with

each other and with other degrees of freedom of the system is specified. For the interactions related to the relaxation processes, this equation provides a general form of magnetization damping.

This paper uses the dynamical invariant method introduced by Lewis and Riesenfeld [10]. It extends its applicability to magnetic systems that are in a non-equilibrium state i.e. the various components in the defining Hamiltonian are time-dependent. This is in contrast to the earlier attempts to solve a problem for quasi-adiabatic evolution.

The  $\vec{\mathbf{D}}$ -term in the magnetization equation has been obtained without using any approximation scheme. It is exact, accumulates all effects of the internal interactions on the magnetization precessional motion and can be a starting point for practical calculations. We have evaluated the  $\vec{\mathbf{D}}$ -term in two special cases, the RSI and the spin-spin interactions. For the RSI, which is linear in spin operators, it takes the form of the Gilbert damping term, the damping and radiation parameters being inter-related. For the spin-spin interactions, it is determined by the spin correlation functions, which fulfil an infinite chain of equations. A further analysis of the  $\vec{\mathbf{D}}$ -term requires an approximation scheme to truncate the chain in a consistent approach to higher order calculations.

In our work, we have considered a specific type of the spin-spin interactions, which do not contribute to the magnetization algebra. However, if spin-spin interactions depend explicitly on the external field, the form of the algebra can change. In this case, the total magnetic moment operator becomes nonlinear in  $\hat{\mathcal{M}}_0$ , and this results in the magnetization algebra with an infinite chain of commutation relations. The chain has to be truncated in a way consistent with the truncation of the chain of equations for the spin correlation functions in the same approximation scheme.

## Acknowledgments

We wish to thank the Natural Sciences and Engineering Research Council of Canada for financial support. The work of S.P.K is funded by the Korea Research Foundation under Grant No. KRF-2003-041-C20053.

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